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TITLE OF THE INVENTION

Hydrogen purifying apparatus

BACKGROUND OF THE INVENTION

The present invention relates to a hydrogen purifying apparatus for use in fuel cells such as solid polymer electrolyte fuel cell. More specifically, the present invention relates to a hydrogen purifying apparatus for reducing a concentration of carbon monoxide in a reformed gas containing gaseous hydrogen and carbon monoxide.

As the fuel gas for use in fuel cells, a reformed gas is used which can be obtained by reforming a material, such as hydrocarbon including natural gas, alcohol including methanol, or naphtha, with a steam or water vapor. Such reformed gas normally contains byproducts including carbon monoxide and carbon dioxide in addition to hydrogen gas.

Fuel cells which can operate at high temperatures, such as molten carbonate fuel cell, can even utilize carbon monoxide as a fuel. However, for fuel cells with lower operating temperatures, such as phosphoric acid fuel cell and solid polymer electrolyte fuel cell, the presence of a high concentration of carbon monoxide in the reformed gas is poisonous to any platinum group metallic catalyst which is utilized by an electrode of the cell. As a result, such cell can not exert satisfactory electric power generating performance. For the solid polymer electrolyte fuel cell in particular, the electrode catalyst is poisoned with carbon monoxide in a

relatively short time even if the concentration of carbon monoxide in the reformed gas is as low as 50 ppm or so, producing rapid impairment of the electric power generating performance of the cell.

Therefore, carbon monoxide is removed by oxidizing it using a platinum group metallic catalyst after reducing the concentration of carbon monoxide in the reformed gas with a carbon monoxide modifying or metamorphic catalyst.

An example of the method for removing carbon monoxide by oxidation is to oxidize only carbon monoxide selectively at low temperature using a catalyst carrying on the carrier alumina a known activator platinum or rhodium, thereby removing the carbon monoxide (see Japanese Laid-Open Patent Publication No. Hei 5-201702, for example).

As an alternative, there is a method which provides an oxidation catalyst of carbon monoxide on the flow pathway of the reformed gas toward the fuel cell and then introduces open air in order to supply sufficient amounts of oxygen (oxidant) to the reformed gas, thereby effectively oxidizing and removing carbon monoxide (see Japanese Laid-Open Patent Publication No. Hei 9-504901, for example).

According to the methods, the concentration of carbon monoxide in the reformed gas can be reduced to as low as 10 ppm or so which is lower than the poisonous concentration to the electrode catalyst.

However, under practical use conditions, since the concentration of carbon monoxide in the reformed gas

changes as the amount of reformed gas supplied to the fuel cell changes, it is necessary to control the supplying amount of open air as appropriate. However, the oxidation reaction of carbon monoxide in the presence of oxidation catalyst accompanies heat generation (exothermic reaction) and thus alters the temperature of the electrode catalyst when the supplying amount of open air to the reformed gas is varied. There is a problem in the prior art methods that when the catalyst temperature is altered and reaches outside the optimal temperature range of the catalyst activity, oxidation and removal of carbon monoxide becomes unsatisfactory.

Another problem is that excess supply of open air increases the amount of heat generated by the oxidation catalyst and elevates the temperature of the catalyst. Heat generation is concentrated particularly at the catalyst close to the side into which the reformed gas is introduced, producing a high temperature around there in a short time. Since hydrogen is more reactive to the catalyst than carbon monoxide, the oxygen supplied as an oxidant is mostly consumed for oxidizing hydrogen rather than carbon monoxide, if the catalyst has a high temperature. As a result, the catalyst loses the ability to selectively oxidize carbon monoxide.

Under such circumstances, it is essential in the hydrogen purifying apparatus to control the catalyst temperature in a range at which carbon monoxide readily reacts with the catalyst but hydrogen does not. In other words, reduction in the change, particularly elevation of catalyst temperature is required.

The most efficient temperature for oxidizing carbon monoxide is a critical low temperature at which carbon monoxide can react with the catalyst. However, control of the temperature of the oxidation catalyst which selectively oxidizes carbon monoxide by regulating the amount of reformed gas to be supplied to the fuel cell or by cooling the catalyst eventually excretes drastic amounts of carbon monoxide upon only a slight decrease of the temperature. Therefore, the prior art methods required control of the temperature within a range of several to several tens degrees centigrade higher than the critical low temperature, in consideration of the flow rate of the reformed gas and possible changes in the catalyst temperature. As such, the conventional methods have met difficulties considerably in achieving selective and efficient oxidation of carbon monoxide.

Furthermore, the oxygen supplied is constantly consumed for oxidizing hydrogen gas as the fuel while being consumed for oxidizing carbon monoxide. This means that there is a need to reduce the supplying amount of open air to a minimum. However, if the catalyst is elevated in temperature when the oxygen amount in hydrogen gas is insufficient due to down-regulated amounts of open air to be supplied, production of carbon monoxide proceeds due to reaction equilibrium between the carbon dioxide and hydrogen. Therefore, the air to be supplied must be controlled exactly and precisely to a right amount. In order to satisfy the above requirement, the conventional hydrogen purifying apparatus meets a problem that it is inevitably complicated in structure.

In view of the above-mentioned various facts, the object of the present invention is to provide a hydrogen purifying apparatus in which the catalyst for selectively oxidizing carbon monoxide can exert the ability sufficiently and the concentration of carbon monoxide in the reformed gas can be reduced constantly and stably even when the use conditions of the apparatus, such as temperature, amounts of open air to be supplied and amounts of carbon monoxide to be treated are varied.

BRIEF SUMMARY OF THE INVENTION

The present invention relates to a hydrogen purifying apparatus for oxidizing and removing carbon monoxide (CO) in a reformed gas containing CO in addition to a main component of hydrogen gas, comprising a reaction segment having a catalyst layer for oxidizing CO, a reformed gas supplying segment for supplying the reformed gas to the reaction segment via a reformed gas supply pathway, an oxidant gas supplying segment for supplying an oxidant gas on the path of the material gas supply pathway, means for cooling the catalyst layer at the upstream side, and means for heating the catalyst layer at the downstream side.

In a preferred mode of the present invention, the means for cooling the catalyst layer at the upstream side is a water-cooled apparatus and the means for heating the catalyst layer at the downstream side is a heater.

In another preferred mode of the present invention, the upstream side of the catalyst layer is formed by a different catalyst from that of the downstream

side, and the catalyst constituting the downstream side exerts the activity at lower temperature than the catalyst constituting the upstream side.

In a further preferred mode of the present invention, the reaction segment may further comprise two or more catalyst layers. In such case, it is preferable to provide each catalyst layer with an oxidant gas supplying segment.

In a still further preferred mode of the present invention, the catalyst layer carries a catalyst on a carrier having a honeycomb structure or a foamed structure with communicating pores.

The catalyst layer is preferably supported by a metallic material.

The heating means is preferably one which utilizes reaction heat generated by reaction of CO and hydrogen in the reformed gas with the oxidant gas.

It is desirable for the catalyst layer that the downstream side has a higher temperature than the upstream side.

In a still further preferred mode of the present invention, the hydrogen purifying apparatus further comprises means for changing the supplying amount of the oxidant gas according to a temperature of the catalyst layer.

In a still further preferred mode of the present invention, the hydrogen purifying apparatus is provided with a flow pathway of the reformed gas at a position close to or in close contact with the catalyst layer via a partition so as to heat said downstream side of said

catalyst layer by a contact with said reformed gas before the passage through the cooling means.

It is desired that the reformed gas passes through the catalyst layer in an opposing direction of stream to that before the same passes through the cooling means.

It is further desired that the reaction segment is placed on the periphery of the flow pathway of the reformed gas before the passage through the catalyst layer.

It is also preferred that the reaction segment is tube-shaped and the flow pathway of the reformed gas before the passage through the cooling means is formed around the reaction segment.

It is also preferable to connect two or more reaction segments in parallel.

In a further preferred mode of the present invention, the hydrogen purifying apparatus is provided with a branched pathway which bifurcates downstream from the connection between the reformed gas supply pathway and the oxidant gas supply pathway and is connected to the reaction segment at the middle point of the catalyst layer, the branched pathway acting as the means for cooling the upstream side of the catalyst layer or otherwise heating the downstream side of the catalyst layer.

In the above case, the hydrogen purifying apparatus preferably further comprises means for changing the cross-sectional area of the reformed gas supply pathway and the branched pathway at their connection to

the reaction segment in order to control the amount of reformed gas to be supplied to the reaction segment from the reformed gas supply pathway and the branched pathway.

It is desirable that the reaction segment has at least a two-segmented catalyst layer and at least the uppermost catalyst layer has a part with no catalytic function or a part with low reactivity to CO.

It is also desirable the reaction segment has at least a two-segmented catalyst layer, and at least the uppermost catalyst layer is composed of a catalyst pellet and the lowermost catalyst layer is in the form of a honeycomb.

It is further desirable that the reaction segment has at least a two-segmented honeycomb shaped catalyst layer, and the uppermost catalyst layer is larger than the lowermost catalyst layer with respect to an open area at the honeycomb lattice.

It is preferred that the uppermost catalyst layer comprises a platinum group metal supported by the alumina group material and the lowermost catalyst layer comprises a platinum group metal supported by the zeolite group material.

The uppermost catalyst layer preferably comprises a catalyst which was heated at a higher temperature than that of the lowermost catalyst layer.

It is preferable for the catalyst layer to have a temperature measuring apparatus and means for controlling the supplying amount of oxidant gas according to a temperature as measured.

While the novel features of the invention are

set forth particularly in the appended claims, the
invention, both as to organization and content, will be
better understood and appreciated, along with other
objects and features thereof, from the following detailed
description taken in conjunction with the drawings.

BRIEF DESCRIPTION OF THE SEVERAL VIEWS OF THE DRAWING

FIG. 1 is a schematic cross-sectional view
illustrating a hydrogen purifying apparatus in the below-
mentioned Embodiment 1-1 of the present invention.

FIG. 2 is a graph illustrating the relation
between the operating temperature of the catalyst for
selectively oxidizing CO and the CO concentration in a
modified gas which has passed through the catalyst layer
obtained from the hydrogen purifying apparatus of FIG. 1.

FIG. 3 is a schematic cross-sectional view
illustrating a hydrogen purifying apparatus in the below-
mentioned Embodiment 1-2 of the present invention.

FIG. 4 is a schematic cross-sectional view
illustrating a hydrogen purifying apparatus in the below-
mentioned Embodiment 1-3 of the present invention.

FIG. 5 is a schematic cross-sectional view
illustrating a hydrogen purifying apparatus in the below-
mentioned Embodiment 1-4 of the present invention.

FIG. 6 is a graph illustrating the operation
characteristics of a catalyst operating at high
temperature and a catalyst operating at low temperature in
a hydrogen purifying apparatus in the below-mentioned
Embodiment 1-4 of the present invention.

FIG. 7 is a schematic cross-sectional view

illustrating a hydrogen purifying apparatus in the below-mentioned Embodiment 1-6 of the present invention.

FIG. 8 is a diagrammatic illustration showing the structure of a hydrogen purifying apparatus embodied in the below-mentioned Embodiment 2-1 of the present invention.

FIG. 9 is a diagrammatic illustration showing the structure of a hydrogen purifying apparatus embodied in the below-mentioned Embodiment 2-2 of the present invention.

FIG. 10 is a diagrammatic illustration showing the structure of a hydrogen purifying apparatus embodied in the below-mentioned Embodiment 2-3 of the present invention.

FIG. 11 is a diagrammatic illustration showing the structure of a hydrogen purifying apparatus embodied in the below-mentioned Embodiment 2-4 of the present invention.

FIG. 12 is a diagrammatical illustration showing the structure of a hydrogen purifying apparatus embodied in the below-mentioned Embodiment 2-5 of the present invention.

FIG. 13 is a diagrammatical illustration showing the structure of a hydrogen purifying apparatus embodied in the below-mentioned Embodiment 2-6 of the present invention.
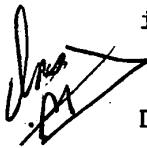
DETAILED DESCRIPTION OF THE INVENTION

The present invention relates to a hydrogen purifying apparatus (or a CO cleanup apparatus) for

oxidizing and removing CO in a reformed gas such as a reformed gas containing CO in addition to a main component of hydrogen gas, comprising a reaction segment having a catalyst layer for oxidizing CO, a reformed gas supplying segment for supplying the reformed gas to the reaction segment via a reformed gas supply pathway, and an oxidant gas supplying segment for supplying an oxidant gas on the path of the reformed gas supply pathway. The most characteristic feature of the present invention is further provisions of means for cooling the catalyst layer at the upstream side, and means for heating the catalyst layer at the downstream side.

The presence of such means facilitates efficient oxidation and removal of undesirable CO in the reformed gas. In other words, such means helps the catalyst which selectively oxidizes CO to exert the ability sufficiently and to reduce CO concentration in the reformed gas (hydrogen gas containing at least CO in this case) stably and constantly even if the use conditions of the apparatus, such as temperature, amounts of open air to be supplied and amounts of CO to be treated are varied.

Here, the means for cooling the upstream side of the catalyst layer and the means for heating the downstream side of the catalyst layer can be embodied in two forms. In the following, Embodiment 1 and Embodiment 2 of the present invention will be described.

Embodiment 1

Embodiment 1 of the present invention uses a water-cooled apparatus as the means for cooling the

upstream side of the catalyst layer and a heater as the means for heating the downstream side of the catalyst layer. The use of such means helps the catalyst for selectively oxidizing only CO to exert the function stably and facilitates control of the catalyst temperature.

In the present embodiment, the means for cooling the upstream side of the catalyst layer is preferably a water-cooled apparatus and the means for heating the downstream side of the catalyst layer is preferably a heater.

It is also preferred that the upstream side of the catalyst layer is formed by a different catalyst from that of the downstream side and that the catalyst constituting the downstream side of the catalyst layer is activated at a temperature lower than the catalyst constituting the upstream side.

The reaction segment may have two or more catalyst layers. In that case, it is preferable to form the oxidant gas supplying segment in each catalyst layer.

A preferred configuration of the catalyst layer is to carry the catalyst on a honeycomb base or a foamed base having communicating pores. In other words, the catalyst may be supported by such honeycomb base or a foamed base having communicating pores.

The catalyst layer is preferably composed of a metallic base carrying the catalyst thereon.

The heating means is preferably such one that utilizes reaction heat generated by the reaction of CO and hydrogen in the reformed gas with the oxidant gas.

It is also preferable to set the temperature of

the downstream side of the catalyst layer higher than that
of the upstream side.

It is preferable to further provide means for
changing the amount of oxidant gas to be supplied in
correspondence with the temperature of the catalyst layer.

The hydrogen purifying apparatus in accordance
with the present invention is preferably provided with a
flow pathway of the reformed gas at a position close to or
in close contact with the catalyst layer via a partition
so as to heat said downstream side of said catalyst layer
by a contact with the reformed gas before the passage
through the cooling means.

It is desired that the reformed gas passes
through the catalyst layer in an opposing direction of
stream to that before it passes through the cooling means.

It is further desired that the reaction segment
is formed on the periphery of the flow pathway of the
reformed gas before the passage through the catalyst
layer.

It is also preferred that the reaction segment
is tube-shaped and the flow pathway of the reformed gas
before the passage through the cooling means is formed
around the reaction segment.

It is also preferable to connect two or more
reaction segments in parallel.

Embodiment 1-1

FIG. 1 is a schematic cross-sectional view of
the hydrogen purifying apparatus in Embodiment 1-1 in
accordance with the present invention. As shown in FIG.

1, the hydrogen purifying apparatus comprises a catalyst layer 1, a reformed gas inlet 2, an air supply duct 3, an air flow rate control valve 4, a cooled water supply duct 5, a cooled water volume control valve 6, a heat exchanger 7, a heater 8, a reaction chamber (or reaction segment) 9, and a reformed gas outlet 10.

In the following, the operation and the technical feature of the present embodiment will be described.

When a fuel is reformed with a water vapor, the CO concentration in the resultant reformed gas increases as the reaction temperature is elevated. When the fuel is methanol which can be reformed at 300 °C or lower, the resultant reformed gas has a CO concentration of 1% or so. Therefore, the reformed gas can be directly introduced through the reformed gas inlet 2. To the contrary, when the fuel is methane or natural gas both requiring 600 °C or so for causing reaction, the resultant reformed gas inevitably contains 10% CO or more due to equilibrium of aqueous gas shift reaction. Therefore, such reformed gas is reduced in the concentration of CO to 1% or less by reacting the same with water vapor using a modifying catalyst to convert CO to carbon dioxide and hydrogen before the same is introduced into the apparatus through the reformed gas inlet 2.

In order to oxidize the CO in the reformed gas, open air is introduced from the air supply duct 3 to cause reaction between the CO and air at the catalyst layer 1. If the introduced air is too little, CO can not react with air sufficiently. If the introduced air is excessive,

hydrogen consumption increases. Therefore, the air volume
to be supplied is regulated by the air flow rate control
valve 4 such that oxygen is contained one to threefold the
amount of CO (volume ratio). The reformed gas mixed with
air is cooled with the heat exchanger 7 down to a
temperature at which the activating temperature of the
catalyst, that is, the catalytic activity for CO of the
catalyst starts to decrease rapidly, or even lower than
that temperature. Then, the reformed gas is supplied to
the catalyst layer 1.

The catalyst layer 1 used here is a ceramic
honeycomb base coated with alumina carrying thereon
platinum. The catalyst layer 1 exhibits the
characteristics as shown in FIG. 2. Namely, since CO in
the reformed gas becomes reactive at a lower temperature
than hydrogen does, the lower the catalyst temperature,
the lower the concentration of CO to be exhausted. If the
temperature is lowered excessively, the temperature
becomes lower than the activating temperature of the
catalyst. As a result, CO becomes nonreactive, which
results in an abrupt increase of CO to be exhausted.

The catalyst layer 1 has been controlled such
that the upstream side has a temperature around or lower
than an activating temperature of the catalyst as used
using the cooling effect of the heat exchanger 7, and the
temperature increases gradually toward downstream. This
structure enables formation of a temperature zone at which
CO is most efficiently reacted with the catalyst layer.
In this way, the CO concentration in the modified gas can
be reduced to the extent of as low as several ppm which is

a level not impairing the performance of any solid polymer electrolyte fuel cell. Any change in the temperature of the reformed gas or the cooling apparatus more or less would result in a mere shift of the position of the above-mentioned temperature zone toward upstream or downstream and would not interfere with stable removal of CO.

In the present embodiment, although open air was used as the oxidant gas, the use of pure oxygen alleviates the decrease of hydrogen concentration in the reformed gas by the amount of nitrogen in open air, because pure oxygen contains no nitrogen. This increases the electric power generating efficiency of the fuel cell which will be connected to the hydrogen purifying apparatus subsequently. However, the use of open air is generally cost-effective.

Similarly, in the present embodiment, although an electric heater was used for heating the catalyst layer 1, oxidation heat generated by CO and hydrogen may be used for the means for heating the downstream side of the catalyst layer 1. By controlling the flow rate of the reformed gas passing through the catalyst layer 1 and the ratio of air to be introduced and by utilizing oxidation heat of CO and hydrogen, the temperature distribution on the catalyst layer 1 can be optimized without use of an electric heater or with the use of minimal heat by a heater.

CO and hydrogen produce more oxidation heat in proportion to the amount of supplied air. Therefore, regulation of air volume can retain a suitable temperature of the catalyst layer 1. This method enables more stable

function of the catalyst layer 1.

In the present embodiment, although the base of the catalyst layer 1 was a codierite honeycomb, any metallic base may also be used. The use of such metallic base for the base of the catalyst layer enables rapid escape of reaction heat, thus facilitating reduction of temperature rises due to oxidation heat by CO and hydrogen. As a result, treated amounts of reformed gas per unit volume by the catalyst layer 1 can be increased and the adverse influence of increases or decreases of the reaction heat due to changes in load on the hydrogen purifying apparatus can be alleviated, leading to stable performance of the apparatus.

Embodiment 1-2

The hydrogen purifying apparatus of Embodiment 1-2 in accordance with the present invention will be described herein. As shown in FIG. 3, the hydrogen purifying apparatus in accordance with the present embodiment comprises a honeycomb catalyst layer 11 formed inside a tube-shaped reaction chamber 18, a reformed gas flow pathway formed external to the reaction chamber 18, and a heat exchange fin 20 provided on a wall of the reformed gas flow pathway neighboring the downstream side of the catalyst layer 11. The operation and effect of the apparatus of this embodiment are mostly similar to those of the apparatus of Embodiment 1-1. Therefore, the description of this embodiment will be focused on different features from those of Embodiment 1-1.

FIG. 3 is a schematic cross-sectional view

illustrating the hydrogen purifying apparatus in Embodiment 1-2. A provision of the heat exchange fin 20 close to the side wall around the downstream side of the catalyst layer 11 helps to heat the downstream side of the catalyst layer 11. Such structure also facilitates cooling the reformed gas by a heat exchanger 17. Moreover, since the reformed gas flow pathway thermally insulates the catalyst layer 11, the temperature distribution in the center and the periphery of the catalyst layer 11 becomes homogeneous, thereby enabling efficient oxidation of CO. Due to the structure of the apparatus such that the reformed gas passes through the catalyst layer 11 in an opposing direction of stream to that before passing through the heat exchanger 17, the reformed gas at elevated temperature can exchange heat with the downstream side of the catalyst layer 11 and is cooled. Because the reformed gas thus cooled then passes along the upstream side of the catalyst layer 11, the temperature of the catalyst layer 11 can be lowered at the upstream side and elevated at the downstream side. As a result, the temperature distribution can be optimized in response to selective oxidation of CO by the catalyst.

In the present example, although the reaction chamber 18 had a single chamber, it can have two or more chambers. In other words, two or more reaction segments can be employed. Two or more reaction chambers can enhance the efficiency of heat exchange with the reformed gas and can cope with a high capacity hydrogen purifying apparatus.

Embodiment 1-3

The hydrogen purifying apparatus of Embodiment 1-3 in accordance with the present invention will be described herein. As shown in FIG. 4, the hydrogen purifying apparatus in accordance with the present embodiment comprises a reaction chamber 28 formed on the periphery of a tube-shaped reformed gas flow pathway, a honeycomb catalyst layer 21 formed inside the reaction chamber 28, and a heat exchange fin 30 provided on a wall of the reformed gas flow pathway neighboring the downstream side of the catalyst layer 21. The operation and effect of the apparatus of this embodiment are mostly similar to those of the apparatus of Embodiment 1-2. Therefore, the description of this embodiment will be focused on different features from those of Embodiment 1-2.

FIG. 4 is a schematic cross-sectional view illustrating the hydrogen purifying apparatus in Embodiment 1-3. A provision of the heat exchange fin 30 on the wall of the reformed gas flow pathway neighboring the downstream side of the catalyst layer 21 helps to heat the downstream side of the catalyst layer 21 while cooling the reformed gas. Such structure also facilitates cooling by a heat exchanger 27. In the event that the flow rate of the reformed gas is increased, cooling only by the heat exchanger proves insufficient occasionally when the temperature is elevated greatly due to oxidation heat by CO and hydrogen. However, the structure of the apparatus of this embodiment where heat is radiated from the periphery of the reaction chamber 28 enables to minimize

elevation of the temperature of the catalyst layer 21. Therefore, the apparatus can cope with any increases in load due to increased flow rate of the reformed gas.

Embodiment 1-4

The hydrogen purifying apparatus of Embodiment 1-4 in accordance with the present invention will be described herein. As shown in FIG. 5, the hydrogen purifying apparatus in accordance with the present embodiment comprises a honeycomb first catalyst layer 31 formed inside a reaction chamber 40 and a second catalyst layer 32 formed downstream from the first catalyst layer 31. The operation and effect of the apparatus of this embodiment are mostly similar to those of the apparatus of Embodiment 1. Therefore, the description of this embodiment will be focused on different features from those of Embodiment 1.

FIG. 5 is a schematic cross-sectional view illustrating the hydrogen purifying apparatus in Embodiment 1-4. The operable temperature range of the catalyst selectively oxidizing CO varies depending on the species of rare metal contained in the catalyst, type of carrier and the like. In the present embodiment, for the first catalyst layer 31, a catalyst operable at high temperatures is used and for the second catalyst layer 32, a catalyst operable at low temperatures. More specifically, a platinum-carrying zeolite was used for the first catalyst layer 31 and a platinum-carrying alumina for the second catalyst layer 32.

As shown in FIG. 6, the catalyst operable at

high temperatures for the first catalyst layer 31 selectively oxidizes only CO at a high success rate when the temperature is high and does not react with CO when the temperature is low, thus allowing non-reacted oxygen to pass away together with CO. Therefore, when the temperature is high, any oxygen is not supplied to the second catalyst layer 32 and therefore the second catalyst layer 32 can play no role in the reaction. To the contrary, when the temperature is low, since oxygen can pass through the first catalyst layer 31 together with CO, the second catalyst layer 32 can play a major role in the reaction thereby removing CO. Moreover, the reaction heat generated at the second catalyst layer 32 heats the downstream side of the first catalyst layer 31, optimizing the temperature distribution of the first catalyst layer 31. As such, arranging two or more catalyst layers each operating in a different temperature range helps to derive catalytic functions of the catalyst layers in a wide range of temperature.

In the present embodiment, although two honeycomb catalyst layers were arranged, three or more catalyst layers may be arranged. Arrangement of many catalyst layers may help to remove CO efficiently in a wide range of temperature.

Alternatively, the catalyst layer may be unitary integrated without dividing it into the first and the second catalyst layers 31 and 32. Integration of the catalyst layer facilitates the installation in the hydrogen purifying apparatus particularly when the catalyst layer comprises a pellet.

Alternatively, the second catalyst layer 32 may use a catalyst which catalyzes the reaction between CO and hydrogen to form methane. When the temperature is controlled at a temperature at which the first catalyst layer 31 consumes all oxygen as supplied, the remaining CO can not be oxidized at the second catalyst layer 32 due to depletion of oxygen. The use of the above-mentioned catalyst forming methane from CO helps to cause reaction between CO and hydrogen, thereby converting CO to methane. The methane forming reaction of CO is prone to proceed at higher temperature than the selective CO oxidation reaction. Therefore, the use of a heater 39 for heating the downstream side of the second catalyst layer as embodied in the present embodiment is preferable.

Embodiment 1-5
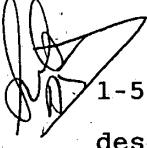
The hydrogen purifying apparatus of Embodiment 1-5 in accordance with the present invention will be described herein. As shown in FIG. 7, the hydrogen purifying apparatus in accordance with the present embodiment comprises a honeycomb first catalyst layer 51 and a second catalyst layer 52 formed downstream from the first catalyst layer 51 which are formed inside a reaction chamber 62, wherein a second air supply duct 55 is provided between the first and the second catalyst layers 51 and 52. The operation and effect of the apparatus of this embodiment are mostly similar to those of the apparatus of Embodiment 1. Therefore, the description of this embodiment will be focused on different features from

those of Embodiment 1.

FIG. 7 is a schematic cross-sectional view illustrating the hydrogen purifying apparatus in Embodiment 1-5. In the present embodiment, for the first catalyst layer 51, a catalyst operable at low temperatures is used and for the second catalyst layer 52, a catalyst operable at high temperatures is used. When the first catalyst layer 51 is increased in temperature greatly due to reaction between CO and air, the catalyst loses the ability to selectively oxidize CO occasionally. Therefore, air supply from a first air supply duct 54 to the first catalyst layer 51 is reduced by providing the second catalyst layer 52 and the second air supply duct 55 in order to suppress temperature rises at the first catalyst layer 51. As a result, CO can be removed efficiently.

Most CO can be removed at the first catalyst layer 51. However, reduced air supply increases slightly the CO concentration to be exhausted compared to the case of no reduction of air supply. Therefore, air corresponding to the volume of remaining CO in the second catalyst layer 52 is supplied to remove the remaining CO. In the structure of this embodiment, since the first catalyst layer 51 is increased in the ability of selective oxidation of CO by the reduction of temperature rises at the first catalyst layer 51, total amounts of air required by the first and the second catalyst layers 51 and 52 will decrease. Moreover, since the amount of air containing oxygen to be supplied to the second catalyst layer 52 is small and the temperature rise due to oxidation heat of CO

is small, the second catalyst layer 52 is heated with a heater 61 in order to retain an optimal temperature of the second catalyst layer 52. In this way, CO can be removed stably and efficiently.

In the present embodiment, the first catalyst layer 51 was formed with the catalyst operating at low temperature and the second catalyst layer 52 with the catalyst operating at high temperature. However, this may be reversed. The use of the catalyst operating at high temperature for the first catalyst layer 51 reduces cooling by a heat exchanger 60. Moreover, the use of the catalyst operating at low temperature for the second catalyst layer 52 enables efficient removal of CO even if the temperature decrease is great due to heat radiation on the way from the first toward the second catalyst layers 51 and 52.

In the foregoing embodiments, Embodiment 1 of the present invention has been described. In the foregoing embodiments, although the hydrogen purifying apparatus used a reformed methane gas and a reformed methanol gas, the present invention is not limited to these gases and can encompass the below-mentioned examples.

In the present mode of embodiment, any reformed gas obtained from other gaseous hydrocarbon fuel, such as propane or butane, and liquid hydrocarbon fuel, such as gasoline or kerosene may be used.

In the foregoing embodiments, although the catalyst carrying platinum on an alumina carrier was mainly used for the catalyst layer, any catalyst can be

used similarly if it can selectively oxidize CO.
Applicable examples are noble metallic catalysts such as
rhodium, ruthenium and composites of these catalysts, and
transition metal composite oxide catalyst having a
perovskite structure. Moreover, silica alumina or zeolite
may be used in place of alumina as the carrier. If
occasion demands, a catalyst which forms methane from CO
selectively may be used.

In the foregoing embodiments, although the
catalyst layer was a honeycomb, any shape such as
spherical shape or pellet may be used similarly if only it
allows the reformed gas to efficiently contact the
catalyst and is less in increases of pressure loss.

In the foregoing embodiments, although the
reformed gas was cooled by means of heat exchange while
circulating water, cooling may be done by circulating an
oil substance such as ethylene glycol having a high
boiling point or a mixed solution of such oil, if
necessary, depending on the temperature. Air to be
supplied to the area where gas modification is carried out
may be used to preheat the reformed material gas.

Embodiment 2

In Embodiment 2 of the present invention, a
branched pathway which bifurcates downstream from the
connection between the reformed gas supply pathway and the
oxidant gas supplying segment and is connected to the
reaction segment at the middle point of the catalyst layer
and said branched pathway acts as said means for cooling
the upstream side of the catalyst layer or otherwise for

heating the downstream side of the catalyst layer.

More specifically, a branched pathway is formed
to vary the amount of oxidant gas to be supplied to the
upstream side and the downstream side of the catalyst
layer in the reaction segment. This structure enables
control of the oxidation reaction of CO which generates
heat as appropriate. In other words, this structure
reduces temperature rises of the catalyst and prevents
development of catalyst reaction in the absence of oxygen.

The hydrogen purifying apparatus having such
structure is advantageous because it can be simplified by
controlling the branched gas volume based on a ratio of
the cross-sectional area of the reformed gas supply
pathway to that of the branched pathway.

The hydrogen purifying apparatus of Embodiment 2
in accordance with the present invention preferably
comprises means for changing the cross-sectional areas of
both the reformed gas supply pathway and the branched
pathway at their connection to the reaction segment in
order to control the volume of the reformed gas to be
introduced into the reaction segment from the reformed gas
supply pathway and the branched pathway.

The reaction segment may have at least a two-
segmented catalyst layer, wherein at least the uppermost
catalyst layer has a part inoperable as a catalyst or a
part with low reactivity to CO.

Alternatively, the reaction segment may have at
least a two-segmented catalyst layer, wherein at least the
uppermost catalyst layer is composed of a catalyst pellet
and the lowermost catalyst layer is shaped into a

honeycomb.

Alternatively, the reaction segment may have at least a two-segmented honeycomb catalyst layer, wherein the uppermost catalyst layer may be larger than the lowermost catalyst layer with respect to an open area of their honeycomb lattice.

It is also preferable that the uppermost catalyst layer comprises a platinum group metal supported by an alumina group material and the lowermost catalyst layer comprises a platinum group metal supported by a zeolite group material.

It is also desirable that the catalyst comprising the uppermost catalyst layer is heated at a higher temperature than that of the catalyst comprising the lowermost catalyst layer.

The catalyst layer is preferred to have a temperature measuring segment and means for controlling an amount of oxidant gas to be supplied according to the temperature as measured.

As shown above, the hydrogen purifying apparatus in accordance with Embodiment 2 of the present invention is an apparatus for oxidizing and removing CO in a reformed gas containing CO in addition to a main component of hydrogen gas, comprising a reaction segment having a catalyst layer for oxidizing CO, a reformed gas supplying segment for supplying the reformed gas to the reaction segment via a reformed gas supply pathway, an oxidant gas supplying segment for supplying an oxidant gas to the reformed gas on the path of the reformed gas supply pathway, and a branched pathway which bifurcates

downstream from a connection between the reformed gas
supply pathway and the oxidant gas supplying segment and
is connected to the reaction segment at the middle of the
catalyst layer. The branched pathway functions as means
for cooling the catalyst layer at the upstream side or
otherwise heating it at the downstream side.

When the reaction segment has a single catalyst
layer, two or more reaction segments may be connected and
a branched pathway may be formed on the reformed gas
supply pathway to connect thereto not only the upstream
reaction segment but also the downstream reaction segment.

In another mode of the present invention, the
hydrogen purifying apparatus comprises a catalyst reaction
segment arranged with a multi-segmented catalyst layer, a
reformed gas supplying segment connected to the catalyst
reaction segment via a reformed gas supply pathway, and an
oxidant gas supplying segment connected halfway to the
reformed gas supplying pathway. And, the apparatus is
characterized in that a catalyst layer with reduced
reactivity to CO is arranged on a first segment so as to
avoid consumption of all oxygen mixed with the reformed
gas by the first segment in order to enable a supply of
oxygen as far as a last or lowermost segment.

In this structure, it is better to form a part
in the first segment which does not function as a
catalyst, that is, a part which allows the reformed gas
mixed with oxygen to pass away without reacting thereto,
or a part which is reactive more or less but low in
reactivity to CO contained in the reformed gas.

The part which does not have catalytic function

may be formed on the catalyst layer by providing such a part that carries no catalyst. Otherwise, after forming the catalyst layer with a carrier carrying a catalyst and a support for supporting the carrier, an opening is formed on the support.

The part with low reactivity to CO may be formed on the catalyst layer by providing a part carrying a reduced amount of catalyst.

Reactivity of the catalyst layer to CO decreases with decreases in surface area of the catalyst layer. Therefore, it is better to form the first segment using a catalyst pellet and the lowermost segment using a honeycomb.

When the catalyst layer is a honeycomb, the larger an open area of honeycomb lattice, the greater the reduction in reactivity of the catalyst layer to CO. Therefore, it is better that the first segment is formed with a catalyst layer which is larger in open area of honeycomb lattice than that of the lowermost segment.

In the hydrogen purifying apparatus of the above-mentioned structure, it is advantageous to form the downstream segment with a catalyst layer which exerts the catalytic activity at higher temperatures than the first segment because this results in stable CO cleanup performance.

This is because a provision of a catalyst layer which exerts the catalytic activity in the same range of temperature as the first segment downstream from the first segment reduces the ability of the downstream segment to selectively oxidize CO, unless the reformed gas entering

the downstream segment is cooled.

Therefore, it is better to use a catalyst layer
carrying a platinum group metallic catalyst on an alumina
carrier for the first segment and a catalyst layer
carrying a platinum group metallic catalyst on a zeolite
carrier which exerts the catalytic activity in higher
temperatures than the first segment for the terminal
segment.

It is also better to use a catalyst layer which
has been heat-treated at a higher temperature than the
terminal segment for the first segment because any
catalyst which has been heated at high temperature is
reduced in the active points and the active range of
temperature is lowered. For example, when the catalyst
layer carries platinum on an alumina carrier, it is better
to use a catalyst layer which has been heat-treated at 700
°C to 800 °C for the first segment and a catalyst layer
which has been heated at 500 °C or so for the terminal
segment.

Furthermore, controlling the volume of oxidant
gas to be supplied based on the temperature of the
catalyst layer as measured by the temperature measuring
segment connected to the catalyst layer is effective for
regulating elevation in temperature of the catalyst layer.

In the following, Embodiment 2 in accordance
with the present invention will be described referring to
the drawings.

Embodiment 2-1

FIG. 8 illustrates the structure of the

hydrogen purifying apparatus in Embodiment 2-1.

A catalyst reaction segment 103 is connected to a material gas supply pathway 105 connected to a material gas supplying segment 101, a material gas exhaust duct 109 connected to a fuel cell, and a branched pathway 108.

The catalyst reaction segment 103 accommodates a first catalyst layer 103a and a second catalyst layer 103b upstream from the gas flow toward downstream in this order. A temperature measuring segment 104a for measuring and indicating a temperature of the first catalyst layer 103a and a temperature measuring segment 104b for measuring and indicating a temperature of the second catalyst layer 103b are also accommodated.

Each of the first and the second catalyst layers 103a and 103b is composed of a catalyst layer carrying, for example, a platinum group metallic catalyst on a honeycomb carrier. A column filled with a platinum group metallic catalyst shaped into a pellet may also be used.

The branched pathway 108 is open into the catalyst reaction segment 103 downstream from the first catalyst layer 103a and upstream from the second catalyst layer 103b.

A joint 106 for supplying air entering from an oxidant gas supplying segment 102 is formed on the path of the material gas supply pathway 105. A branched volume control segment 107 connected to the branched pathway 108 is formed between the joint 106 and the catalyst reaction segment 103 to control the volume of material gas to be passed to the branched pathway 108.

The material gas supplying segment 101 has an

apparatus for supplying a reformed (material) gas which supplies hydrogen gas and contains at least a byproduct CO. This supplying apparatus is a reactor which reforms, for example, hydrocarbon material gas, alcohol or naphtha with steam to generate hydrogen.

An apparatus for supplying an oxidant gas containing at least oxygen is installed inside the oxidant gas supplying segment 2. This apparatus may be exemplified as air pump supplying air as the oxidant gas, oxygen bomb, etc.

Next, operations of the hydrogen purifying apparatus in accordance with Embodiment 2 of the present invention will be described.

A material gas which has been generated in the material gas supplying segment 101 is supplied to the catalyst reaction segment 103 through the material gas supply pathway 105 while supplying air from the oxidant gas supplying segment 102 through the joint 106 at the same time. At that time, the volume (mole) of oxygen contained in the air may be adjusted to, for example, half the volume of CO in the material gas.

Some part of the material gas mixed with oxygen enters the upstream side of the first catalyst layer 103a in the catalyst reaction segment 103 through the material gas supply pathway 105. The remaining part enters the downstream side of the first catalyst layer 103a through the branched pathway 108 and mixed with the material gas which has passed through the first catalyst layer 103a. The volume of the material gas entering the material gas supply pathway 105 and the branched pathway 108 is

regulated by the branched volume control segment 107.

Then, the material gas mixture is passed to the second catalyst layer 103b to oxidize and remove CO in the material gas there. The material gas from which CO has been removed and cleaned up is supplied to the fuel cell through the material gas exhaust duct 109.

As such, by branching the material gas mixed with oxygen to clean up some part of the material gas at the first catalyst layer, the amount of heat generating at the first catalyst layer can be reduced.

Furthermore, since the material gas which avoids the first catalyst layer is passed to the second catalyst layer, temperature rise in the material gas entering the second catalyst layer can be prevented. This in turn suppresses temperature rise in the second catalyst layer. Furthermore, this structure facilitates supply of oxygen necessary for cleaning up the material gas by removing CO at the second catalyst layer thereby preventing depletion of oxygen during catalyst reaction at the second catalyst layer.

As discussed above, the present embodiment allows the catalyst which selectively oxidizes CO to exert the catalytic activity sufficiently and facilitates removal of more amounts of CO, that is, cleanup of the material gas flowing at a high rate.

Furthermore, since this embodiment permits the monistic control of the volume of air to be supplied, an exact volume of air can be supplied and thus consumption of hydrogen gas by excess oxygen can be prevented.

The volume of material gas passing through the

upstream side of the first catalyst layer 103a and that passing through the branched pathway 108 may be controlled based on a ratio of the cross-sectional area of the material gas supply pathway 105 to that of the branched pathway 108 without forming the branched volume control segment 107. For example, when the ratio is set at 1:1, the material gas is branched in half.

The above structure facilitates simplified control of the catalyst layer in an optimal range of temperature.

This structure also facilitates control of the catalyst layer temperature with high accuracy by adjusting the branched volume of material gas based on the temperature measured by the catalyst layer temperature measuring segment 104. This is advantageous in cleaning up CO in correspondence with a change in the flow rate of hydrogen.

An oxygen volume (mole) in the air to be supplied less than half the volume of CO in the material gas is more likely to lead to depletion of oxygen. Therefore, it is better to determine the air volume to be supplied to make an oxygen volume more than half the volume of CO.

Next, a specific example of operation of the hydrogen purifying apparatus in accordance with Embodiment 2 will be described.

A steam reforming apparatus for natural gas is used as a material gas generator of the material gas supplying segment 101. A material gas containing about 80% hydrogen, about 19.5% carbon dioxide and about 0.5%

carbon monoxide on the basis of dry gas was generated. The resultant material gas was passed to the material gas supply pathway 105 at a rate of 10 1/min.

Air containing oxygen about twice the volume of CO was supplied to the material gas through the joint 106 using an air pump installed at the oxidant gas supplying segment 102.

After mixed with air, the material gas was branched in about half by the branched volume control segment 107. The branched half was passed to the upstream side of the first catalyst layer 103a in the catalyst reaction segment 103.

Gas analysis of the material gas collected after it had passed through the first catalyst layer 103a indicated that the material gas contained 10 ppm CO or less. Similar analysis of the material gas collected after it was mixed with the branched material gas indicated that the material gas mixture contained about 0.25% CO and oxygen about twice the volume of CO.

Gas analysis of the material gas excreted from the material gas exhaust duct 109 revealed the presence of 10 ppm CO or less in the material gas.

A doubling of the volume of material gas to be supplied also decreased the CO concentration in the material gas to as low as 10 ppm or less.

Embodiment 2-2

FIG. 9 illustrates the structure of the hydrogen purifying apparatus in accordance with Embodiment 2-2 of the present invention.

A first catalyst reaction segment 111 is formed upstream from the material gas stream and a second catalyst reaction segment 114 downstream from the material gas stream. The first and the second reaction segments are connected to each other via a communicating pathway 113.

While the first catalyst reaction segment 111 is connected to a material gas supply pathway 105 which is connected to the material gas supplying segment 101, and the second catalyst reaction segment 114 is connected to a material gas exhaust duct 109 which is connected to a fuel cell. A branched pathway 108 bifurcating from the material gas supply pathway 105 is connected to the communicating pathway 113 at a junction 110.

A first catalyst layer 111a and a temperature measuring segment 112 for measuring the temperature of the first catalyst layer 111a are formed in the first catalyst reaction segment 111. Similarly, a second catalyst layer 114a and a temperature measuring segment 115 for measuring the temperature of the second catalyst layer 114a are formed in the second catalyst reaction segment 114. The remaining parts are formed in the same manner as in Embodiment 2-1.

A division of the catalyst reaction segment into two parts facilitates effectively mixing the material gas which has passed through the first reaction segment with the material gas which is supplied through the branched pathway 108. This structure also facilitates effectively cooling the material gas which has been heated at the first catalyst reaction segment whereby cleanup of CO can

be performed stably.

Embodiment 2-3

FIG. 10 shows the structure of the hydrogen purifying apparatus in accordance with Embodiment 2-3 of the present invention.

A catalyst reaction segment 116 is connected not only to a material gas supply pathway 105 which is connected to a material gas supplying segment 101 but also to a material gas exhaust duct 109 which is connected to a fuel cell.

The catalyst reaction segment 116 accommodates therein a first catalyst layer 116a and a second catalyst 116b upstream from the catalyst reaction segment 116 toward downstream in this order. The catalyst reaction segment 116 also accommodates a temperature measuring segment 119a for measuring and indicating the temperature of the first catalyst layer 116a and a temperature measuring segment 119b for measuring and indicating the temperature of the second catalyst layer 116b. The first catalyst layer 116a has a catalyst-free part 116c at the periphery. A joint 106 for supplying air from an oxidant gas supplying segment 102 is formed on the path of the material gas supply pathway 105. Identical apparatuses to those of Embodiment 2-1 are installed in the material gas supplying segment 101 and the oxidant gas supplying segment 102.

In the present embodiment, the material gas is not branched after it is supplied and mixed with air in order to pass all the material gas to the catalyst

reaction segment 116 and the first catalyst layer has the catalyst-free part 116c.

Since oxidation reaction does not occur at the catalyst-free part 116c, the material gas which has passed through this part is not elevated in temperature. Moreover, no oxygen is consumed at this part. Therefore, sufficient and stable cleanup of CO can be performed at the second catalyst layer 116b.

Omission of a branched pathway on the flow pathway of material gas which has been supplied and mixed with the oxidant gas simplifies the structure of the resultant hydrogen purifying apparatus.

The position of the catalyst-free part in the catalyst layer is not limited to the periphery of the catalyst layer and it may be positioned at any site in the catalyst layer, such as center, for example.

The volume of material gas which has not been treated and is to be passed to the second catalyst layer can be regulated based on a ratio of area of the catalyst-carrying part of the catalyst layer to that of the catalyst-free part.

For example, when the ratio is 1:1 to 1:10 or so, cleanup of CO can be performed stably.

In the following, an example of concrete operations of the hydrogen purifying apparatus of this embodiment will be described.

After setting the ratio of the area of the catalyst-carrying part 116a to that of the catalyst-free part 116c at 1:1, air was supplied to the material gas generated in the same manner as in Embodiment 2-1. The

material gas was passed into the catalyst reaction segment 116 in order to treat the material gas. Gas analysis of the material gas excreted from the material gas exhaust duct 109 indicated that the material gas contained 10 ppm CO or less.

Then, even when the supplied amount of material gas was doubled, a CO concentration of 10 ppm or less could be achieved.

The ratio of the area of the catalyst-carrying part to that of the catalyst-free part should desirably be determined in correspondence with the cleaning up ability of the catalyst used and it is not limited to the range of 1:1 to 1:10.

Embodiment 2-4

FIG. 11 illustrates the structure of the hydrogen purifying apparatus in accordance with Embodiment 2-4 of the present invention.

A catalyst reaction segment 117 is connected to both a material gas supply pathway 105 which is connected to a material gas supplying segment 101 and a material gas exhaust pathway 109 which is connected to a fuel cell.

The catalyst reaction segment 117 accommodates a first catalyst layer and a second catalyst layer 117b upstream from the catalyst reaction segment to downward in this order. The first catalyst layer is composed of a catalyst carrying part 117a which carries a platinum group metallic catalyst and a support 117c formed on the periphery of the catalyst carrying part 117a. The support has an air vent hole 117d. The catalyst reaction segment

117 also accommodates a temperature measuring segment 120a for measuring and indicating the temperature of the catalyst carrying part 117a and a temperature measuring segment 120b for measuring and indicating the temperature of the second catalyst layer 117b. The remaining parts are arranged in the same manner as in Embodiment 2-3.

Since the material gas which has passed through the air vent hole 117d is not elevated in temperature and has consumed no oxygen, the second catalyst layer can clean up CO sufficiently.

When a ratio of the total area of the air vent hole 117d to the area of the catalyst carrying part 117a is 1:1 to 1:10 or so, CO cleanup can be done stably; however, the ratio should desirably be determined in correspondence with the cleaning up ability of the catalyst used and is not limited to the range of 1:1 to 1:10.

Embodiment 2-5

FIG. 12 illustrates the structure of the hydrogen purifying apparatus in accordance with Embodiment 2-5 of the present invention.

A catalyst reaction segment 118 is connected to a material gas supply pathway 105 which is connected to a material gas supplying segment 101 and a material gas exhaust duct 109 which is connected to a fuel cell.

The catalyst reaction segment 118 accommodates therein a first catalyst layer 118a and a second catalyst layer 118b upstream from the catalyst reaction segment 118 to downward in this order. The catalyst reaction segment

118 also accommodates a temperature measuring segment 121a
for measuring and indicating the temperature of the first
catalyst layer 118a and a temperature measuring segment
120b for measuring and indicating the temperature of the
second catalyst layer 118b. For the first catalyst layer
118a, a catalyst layer with a less number of honeycomb
lattices per unit area than that of the second catalyst
layer 118b is used. The remaining parts are arranged in
the same manner as in Embodiment 2-3.

The use of such catalyst layer with reduced
reactivity to CO for the first catalyst layer prevents
consumption of all oxygen supplied to the material gas at
the first catalyst layer and thus allows oxygen to be
passed to the second catalyst layer. This structure also
enables suppression of temperature rise at the first
catalyst layer thereby suppressing temperature rise of the
material gas when it passes through the second catalyst
layer. As a result, cleanup of CO can be done
sufficiently at the second catalyst layer.

The first catalyst layer 118a may be formed
using a catalyst layer shaped in a pellet and the second
catalyst layer 118b using a catalyst layer carrying a
catalyst on a honeycomb carrier.

Embodiment 2-6

FIG. 13 illustrates the structure of the
hydrogen purifying apparatus in accordance with Embodiment
2-6 of the present invention.

A catalyst reaction segment 122 is connected to
a material gas supply pathway 105 which is connected to a

material gas supplying segment 101, a material gas exhaust duct 109 which is connected to a fuel cell, and a branch 108.

The catalyst reaction segment 122 accommodates a catalyst layer 123 and a temperature measuring segment 124 for measuring and indicating the temperature of the catalyst layer 123. The catalyst layer 123 is a column filled with a platinum group metallic catalyst shaped in a pellet. The branched pathway 108 is connected to the catalyst reaction segment 122 on a side of the catalyst layer 123. The remaining parts are arranged in the same manner as in Embodiment 2-1.

Such structure reduces heat generation at the catalyst layer 123. Furthermore, since material gas which has been branched on the way to the catalyst layer 123 is supplied to the middle of the catalyst layer 123, CO in the reformed gas can be cleaned up sufficiently.

In the following, examples of Embodiment 1 in accordance with the present invention will be described.

Example 1

An alumina carrying platinum was coated on a codierite honeycomb of 50 mm diameter and 100 mm length to form the catalyst layer 1. The catalyst layer 1 thus formed was placed in the reaction chamber 9 of the hydrogen purifying apparatus as shown in FIG. 1 and then a reformed gas containing 1% CO, 15% carbon dioxide, 15% water vapor and hydrogen for the rest was introduced into the apparatus through the reformed gas inlet 2 at a rate of 10 1/min. Open air was supplied from the air supply

duct at 1 l/min. The temperature of the mixed reformed
gas with air was cooled to 100 °C with the heat exchanger
7 in which water is circulated to cause reaction between
CO and oxygen at the catalyst layer 1. The catalyst layer
1 was heated with the heater 8 at the downstream side to
make a temperature of 150 °C. Measurement of temperature
distribution on the catalyst layer 1 indicated an almost
linear elevation from upstream side toward downstream.
The composition of the reformed gas which was excreted
from the reformed gas outlet 10 after it had passed
through the catalyst layer 1 was measured by gas
chromatography. The result indicated a CO concentration
of 5 ppm. Then, the flow rate of water circulating inside
the heat exchanger 7 was varied to alter the temperature
of the reformed gas to 90 °C and 80 °C. The CO
concentration in the resultant reformed gas was 6 ppm and
9 ppm, respectively. Then, the set temperature of the
heater 8 was varied to adjust the temperature of the
downstream side of the catalyst layer 1 to 140 °C and 160
°C. The CO concentration in the resultant reformed gas
was 4 ppm and 9 ppm, respectively.

Example 2

In this example, the catalyst layer as formed in
the above Example 1 was installed inside the reaction
chamber 18 as shown in FIG. 3 and a reformed gas was
introduced into the apparatus through the reformed gas
inlet 12. Measurement of the temperature of the reformed
gas at the reformed gas inlet 12 showed 200 °C.
Measurement of the temperature of the downstream side of

the catalyst layer 11 which had been heated by the heat
exchange fin 20 showed 150 °C. Then, the reformed gas was
cooled down to 100 °C using the heat exchanger 17 and
reacted at the catalyst layer 11. This produced a CO
concentration of 5 ppm.

Example 3

As shown in FIG. 4, a codierite honeycomb base
of 100 mm diameter and 100 mm length having a punched hole
of 80 mm diameter in the center was coated with an
identical catalyst to that in Example 1 to form the
catalyst layer 21. Measurement of the temperature of the
downstream side of the catalyst layer 11 which had been
heated by the heat exchange fin 30 showed 150 °C. Then,
the reformed gas was cooled down to 100 °C using the heat
exchanger 27 and reacted at the catalyst layer 21. This
produced a CO concentration of 4 ppm.

Example 4

A codierite honeycomb of 50 mm diameter and 50
mm length was coated with platinum-carrying mordenite to
form the first catalyst layer 31. Similarly, a codierite
honeycomb of the same size was coated with platinum-
carrying alumina to form the second catalyst layer 32.
The first and the second catalyst layers 31 and 32 thus
formed were placed in the reaction chamber 40 as shown in
FIG. 5. Then, the modified gas was cooled down to 140 °C
using the heat exchanger 38. The temperature of the
second catalyst layer 32 was controlled with a heater 39
such that the downstream side had a temperature of 160 °C.

Measurement of the temperature of the downstream side of
the first catalyst layer 31 showed 150 °C. Measurement of
the CO concentration at the reformed gas outlet 41
indicated 1 ppm.

Example 5

        In this example, a codierite honeycomb measuring
50 mm in diameter and 50 mm in length was coated with
platinum-carrying mordenite to form the first catalyst
layer 51. Similarly, a codierite honeycomb of the same
size was coated with platinum-carrying alumina to form the
second catalyst layer 52. Both the first and the second
catalyst layers 51 and 52 were installed in the reaction
chamber 62 as shown in FIG. 7. The reformed gas was
cooled down to 150 °C using the heat exchanger 60. The
temperature of the second catalyst layer 52 was controlled
with the heater 61 such that the downstream side had a
temperature of 130 °C. Measurement of the temperature at
the downstream side of the first catalyst layer 51 and the
upstream side of the second catalyst layer 52 indicated
140 °C for the former and 100 °C for the latter.
Measurement of the CO concentration at the reformed gas
outlet 63 showed 2 ppm.

Comparative Example 1

        After omitting the heater 8 from the apparatus
of Example 1, the modified gas was introduced into the
apparatus through the reformed gas inlet 2 in the same
manner as in Example 1. The reformed gas was cooled down
to 100 °C using the heat exchanger 7 and reacted at the

catalyst layer 1. Measurement of the temperature distribution in the catalyst layer 1 showed a temperature of 100 °C at the upstream side. The temperature showed a linear decrease toward downstream and the temperature was 90 °C at the lowermost. Gas chromatography of the composition of the reformed gas excreted from the reformed gas outlet 10 after it had passed through the catalyst layer 1 indicated a CO concentration of 2,000 ppm. Then, the flow rate of the circulating water in the heat exchanger 7 was changed to vary the temperature of the reformed gas to 90 °C, 110 °C and 120 °C. These temperatures produced CO concentrations of 5,000 ppm, 200 ppm and 500 ppm, respectively.

Comparative Example 2

The heater 61 was omitted from the apparatus of Example 5. Measurement of the temperature at the downstream side of the first catalyst layer 51, and at the upstream side and downstream side of the second catalyst layer 52 indicated 140 °C, 100 °C and 90 °C, respectively. The CO concentration measured at the reformed gas outlet 63 was 500 ppm.

Although the present invention has been described in terms of the presently preferred embodiments, it is to be understood that such disclosure is not to be interpreted as limiting. Various alterations and modifications will no doubt become apparent to those skilled in the art to which the present invention pertains, after having read the above disclosure. Accordingly, it is intended that the appended claims be

interpreted as covering all alterations and modifications as fall within the true spirit and scope of the invention.